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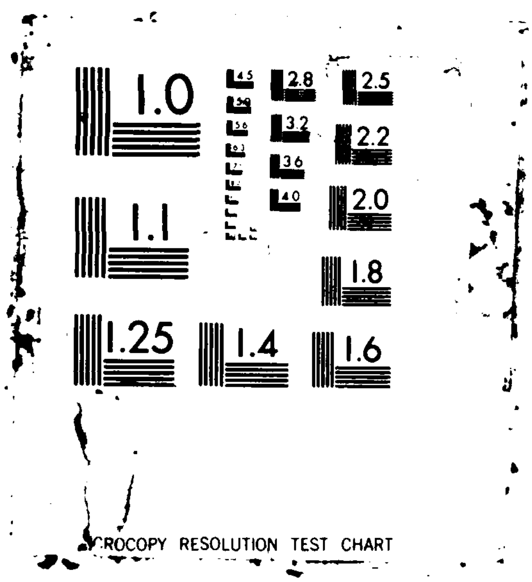
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This annual report summarizes Yale High Temperature Chemical Reaction Engrg. Laboratory research methods/results (Grant AFOSR 84-0034) for the ca one-year period ending 12/31/86. Our techniques and results are outlined in the areas of (1) laser-based real-time optical techniques for measuring soot particle thermophoretic diffusivities, and vapor and/or particle-deposition rates onto cooled surfaces in combustion gases, (2) role of thermophoresis in the capture of soot particles and the use of this phenomenon to infer both local soot volume fractions and local gas temperatures, (3) boundary layer computational methods and correlations for thermophoretically-modified vapor and small particle transport, including high mass-loading and dopant redistribution effects, and, (4) use of a micro-wave-induced plasma emission spectroscopic (MIPES) method to follow boron surface gasification kinetics in streams containing $O_2(g)$. Presentation and archive publications describing these techniques/findings are documented, including the publication of a more comprehensive textbook: Transport Processes in Chemically Reacting Flow Systems (Butterworths, Stoneham MA) by the Principal Investigator. Keywords: oxygen

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1. INTRODUCTION

The performance of ramjets burning slurry fuels (leading to condensed oxide aerosols and liquid film deposits), gas turbine (GT) engines in dusty atmospheres, or when using fuels from non-traditional sources (e.g., shale-, or coal-derived), depends upon the formation and transport of small particles across non-isothermal combustion gas boundary layers (BLs). Moreover, even airbreathing engines burning "clean" hydrocarbon fuels can experience soot formation/deposition problems (e.g., combustor liner burnout, accelerated turbine blade erosion and "hot" corrosion). Accordingly, our research is directed toward providing chemical propulsion systems R & D engineers with new techniques and quantitative information on important particle and vapor mass transport and kinetic mechanisms and rates.

An interactive experimental/theoretical approach is being used to gain an understanding of performance-limiting chemical-, and mass/energy transfer-phenomena at or near interfaces. This includes the development and exploitation of seeded laboratory flat flame burners (Fig. 1) and cooled deposition targets, flow-reactors (Fig. 2), and new optical diagnostic/ spectroscopic techniques. Resulting experimental rate data, together with the predictions of comprehensive asymptotic theories, are then used as the basis for proposing and verifying simple viewpoints and effective engineering correlations for future design/optimization studies.

The purpose of this report is to briefly summarize our research methods and accomplisments under AFOSR Grant 84-0034 (Technical Monitor: J.M. Tishkoff) during the one-year period: 12/1/85 - 12/31/86. Readers interested in greater detail than contained in Section 2 are advised to consult the published papers cited in Sections 2, 5. Copies of any of these published papers or preprints can be obtained by writing the PI: Prof. Daniel E. Rosner at the Department of Chemical Engineering, Yale University, Box 2159 Yale Station, New Haven, CT 06520, U.S.A. Comments on, or examples of, the applicability of our research results will be especially welcome.

2. RESEARCH ACCOMPLISHMENTS AND PUBLICATIONS

Most of the results we have obtained under Grant AFOSR 84-0034 can be divided into the three subsections below:

2.1. Seeded Flame Experiments on Vapor and Submicron Particulate Transport Rates

2.1.1. Coupling Between Particle Deposition (1) and Vapor Deposition (10)

Castillo and Rosner (12) predicted that the simultaneous presence of submicron particulate matter would reduce the rate of vapor deposition on to surfaces cooled below the vapor dew-point temperature. Thus, even though the particles which scavenge the vapor are thermophoreticallly 'attracted' to the cool surface, the net rate of condensate arrival is reduced since the Fick diffusivity of the parent vapors is larger than the particle thermophoretic diffusivity. This prediction (12) has now been verified in the recent seeded flame experiments of Rosner and Liang (20, 21), carried out using submicron MgO(s) particles and Na_2SO_4(g) vapor by combining the techniques of flash evaporation (alkali emission spectroscopy) (10, 21) with laser light scattering (Fig. 1). Both the experiments and theory demonstrate that the reduction in condensible species transport rates is primarily a function of the scavenging particle surface area per unit volume within the boundary layer (12, 26).

2.1.2. Thermophoretic Inference of Local Soot Volume Fraction and Gas Temperature in Soot-Laden Combustion Gases (5).

As is well known, the inference of accurate local gas temperature in soot-laden flame regions poses formidable challenges, and has motivated the recent application of coherent anti-Stokes Raman spectroscopic (CARS) methods applied to the ubiquitous species N_2. Despite the equipment and optical access demands of this "non-intrusive" technique, inferred gas temperatures may still be uncertain to about $\pm$ 50K in N_2-rich regions, otherwise + 75/ - 125K. Moreover, C_2(g)-bands produced by pulsed laser vaporization of the soot can interfere with the CARS inference of N_2-temperatures.

In a recent paper (5) we demonstrated that the temperature vs. time response of a thermocouple suddenly immersed in the soot-laden region of a laminar flame can be quantitatively related to the themophoreticlly-dominated soot acquisition rate by the thermocouple. Indeed, the slope of a suitable straight line plot of the recorded thermocouple output was shown to provide quantitative information about local soot volume fraction, f_v, in the gas, irrespective of local particle size distribution, or particle thermal/optical properties in a multi-dimensional laminar flame. In our more recent work, we have emphasized a potentially equally important by-product of this simple technique, viz, the thermophoretically-inferred local gas temperature in a region of the flame which contains soot. Our strategy is that, once our thermocouple response theory is verified and accepted, the value of T_g in a soot-laden region of the flame can be estimated to be that value which, when introduced into our thermophoretic probe theory, produces the best straight line thermocouple response data. Indirectly, this is equivalent to estimating T_g as that (extrapolated) surface temperature at which local thermophoretic particle capture rate would vanish.

Remarkably enough, the presence of soot then becomes the means by which T_g is estimated, not the complication which precludes its accurate inference! Moreover, the resulting T_g should be insensitive to the (inevitable) uncertainty in the thermophoretic diffusivity of soot particles, and, as emphasized ealier (5), the local soot volume fraction, f_v, is also obtained *via* this procedure.

Considering the abovementioned factors, it appears that the phenomenon of soot particle thermophoresis and its unique characteristics may, in the future, be exploited to map out local f_v and T_g combinations in multidimensional laminar flows without requiring the now-feasible but complex alternatives of, say, optical tomography (multi-beam extinction) and CARS.

2.1.3. Experimental Determination of 'Soot' Particle Thermophoretic Diffusivity Using Seeded Opposed Jet Laminar Diffusion Flame Techniques

To determine the thermophoretic diffusivity of submicron aerosol particles in a well controlled temperature and velocity field, we are currently seeding an atmospheric pressure opposed jet counterflow diffusion flame (CH_4+N_2/O_2+N_2) with TiO_2 particles. The particles are generated by the chemical reaction of titanium chloride vapor (evaporated into the oxidizer stream) with water vapor (generated at the flame reaction "sheet"). When the flame is operated at very low strain rates (4 s^{-1}) and the flow rates are adjusted so that the flame is superimposed on the gas stagnation plane, the particle stagnation plane should be measurably displaced with respect to the gas stagnation plane (eg., *ca.* 2mm. when N_2 is the 'carrier' gas). In fact, the particle stagnation plane positions itself where the thermophoretic velocity (in the direction opposite to the temperature gradient) is counterbalanced by the convective velocity of the approaching stream. The thermophoretic diffusivity is then determined by: i) measuring (laser light scattering) the displacement between the two stagnation planes; ii) measuring the local temperature field using a fine thermocouple, and iii) calculating the local gas velocity field using a numerical code.

2.2. Transport Theory: Thermophoretically Modified Boundary Layer Convective Mass Transport

We have extended our previous solutions and correlations of thermophoretically modified submicron particle mass transport across laminar (3,7,8) and turbulent BLs (6,8) into the domain of high particle mass loading (16), a situation encountered in numerous materials processing applications, and, locally, in two-phase (e.g., droplet/gas) flows of chemical propulsion interest. We have also developed methods to predict the extent to which additive vapors, soluble in a particulate phase (eg., dopants to control optical properties of a deposit), are incorporated into deposits under high mass-loading conditions (22). Also, because of increasing interest in the Soret diffusion of large, highly nonspherical molecules and the thermophoretic transport of nonspherical particles (e.g., long soot aggregates) we have recently predicted (*via* gas kinetic theory) their thermal diffusion factors, α_T. These results have now been prepared for publication, and will be submitted in early 1987 (15). We hope to experimentally test some of these predictions, and their BL-consequences, in the future.

Two developments in our convective mass transfer research not especially associated with particle thermophoresis are also noteworthy. One is our recent theoretical treatment of chemical vapor deposition (CVD) rates in each of the two asymptotic kinetic extremes: a) no vapor phase chemistry (the 'chemically frozen' BL limit), b) local equilibrium vapor phase chemistry (the 'local thermochemical equilibrium' (LTCE) limit). Our results (now being prepared for presentation at the next international conference on CVD (23)) reveal that, if LTCE is established at the vapor/solid interface by virtue of *heterogenous* reactions CVD-rates are quite insensitive to the (usually unknown) vapor phase chemical kinetics. Second, during this reporting period the PI's textbook: *Transport Processes in Chemically Reacting Flow Systems* has been published (13). It incorporates many results of our previous and current AFOSR research programs (see author's Preface) and will help educate many engineers / applied scientists in U.S. Universities, and industrial contractor / government laboratories.

2.3. Heterogeneous Kinetics (18, 24)

To make (i) rapid-response gas/solid reaction rate measurements over a large temperature range, and (ii) surface mass balances necessary for mechanistic understanding of high temperature gas/solid reactions, we have recently been exploiting an emission spectroscopic technique. In this technique, a low pressure *microwave-induced plasma* (MIP) excites characteristic emission from the atoms in the gaseous product species of a gas/solid reaction in a low pressure flow reactor.

We employ a modified version of our transonic, vacuum flow reactors developed earlier under AFOSR-support for the study of gas reactions with silicon- and boron-containing refractory solid compounds (18). However, now the reaction *product* vapor species are dissociated and electronic emission from the resulting atoms is produced in a microwave discharge plasma before leaving the reactor (Fig. 2). Evaporation and gasification reactions are studied by measuring emission intensity, I, from this discharge, *via* a 0.5m Jarrell-Ash monochromator.

Aside from steady-state reaction rate measurements, flash evolution experiments can be carried out to measure the amount of condensed product material formed on a surface during reaction, provided, of course, that the reaction product (e.g., B_2O_3) has a higher volatility than that of the substrate, e.g., B(s). In such experiments the filament is exposed to the gaseous reagent for some reaction time (normally only a few minutes). Then, the gaseous reagent flow into the reactor is stopped and the filament cooled. Finally, the I(t) is determined when the filament is heated rapidly.

We are now performing preliminary experiments on the application of this microwave-induced plasma emission spectroscopy (MIPES) technique to the oxidation of boron (24), a system of considerable interest to the propulsion community, but one whose poorly understood kinetics are apparently influenced by the condensibility of the reaction product B_2O_3. Preliminary results have been obtained for the high temperature gasification kinetics of boron by O_2(g), and CO_2(g),and were reported at the last Eastern States Combustion Institute Conference. We are now set up to initiate measurements of the oxidation kinetics of boron by B_2O_3(g) (*i.e.*, OBOBO(g)) and exploit the rapid-response characteristics of our MIPES technique to measure the behavior of such surfaces when their surface temperatures are modulated, or in modulated reactant streams. Among other things, such studies could shed valuable light on the response of solid fuel surfaces in a turbulent environment.

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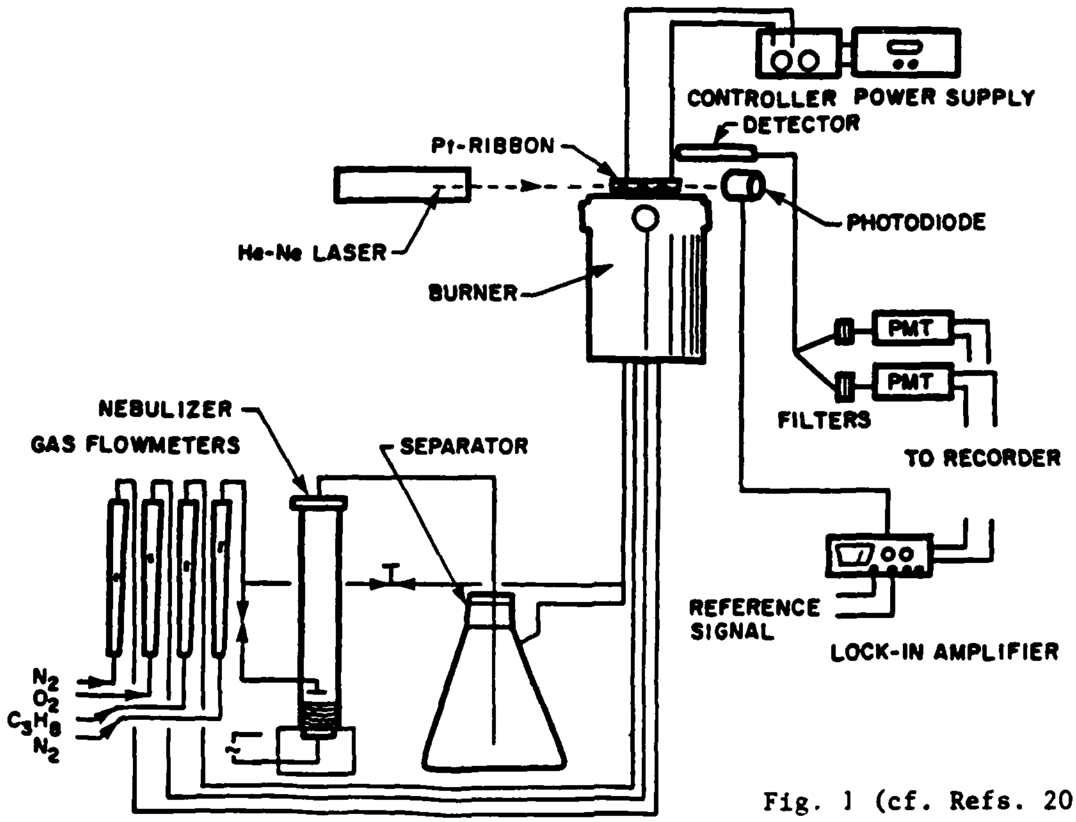

Fig. 1 (cf. Refs. 20, 21)

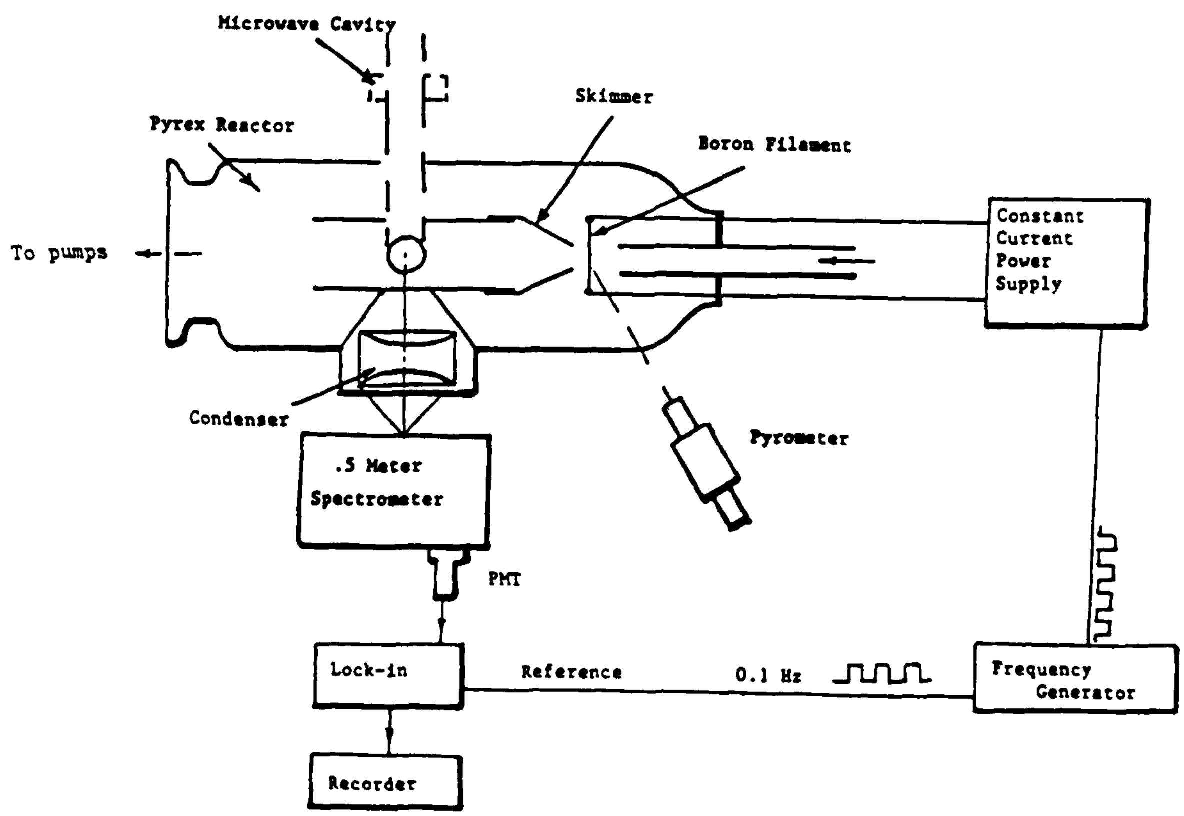

Fig. 2 (cf. Ref. 24)

3. ADMINISTRATIVE INFORMATION ; PERSONNEL AND PRESENTATIONS

Table 3.1 summarizes the personnel who have contributed to this research program during the period: 12/1/85 - 12/31/86, along with the subject matter of each investigators research contribution.

Table 3.1

SUMMARY OF PERSONNEL AND THEIR CONTRIBUTIONS

Name	Status @ Yale	Primary Contribution
Rosner, D.E.	PI[a], ChE	Overall Program Direction[19]
Gomez, A.	PDRA-Lecturer	Measurement of Thermophoretic Properties of Soot Particles
Garcia-Ybarra, P.	PDRA[c]-Visiting Scholar	Thermophoretical Properties of Nonspherical Particles15
Liang, B.	GRA[b] ('87)	Vapor and/or Particle Deposition Interaction[10]
Naragajan, R.	GRA ('86)	Dynamics of C.V.D. Condensate Layers[17]
Park, H.M.	GRA ('87)	Theory of High-Mass-Loaded Aerosol Transport[16,22]
Roy, R.	GRA (MA)	Thermodynamics of Nonideal Condensate Mixtures
Timmins, M.	UGRA[d] (Yale '87)	Measurement of Thermophoretic Properties of Soot Particles
Zvuloni, R.	GRA ('87)	Boron Gasification Kinetics[24]

[a] Principal Investigator
[b] Graduate Research Assistant (Anticipated Year of PhD Degree)
[c] Postdoctoral Research Assistant
[d] Undergraduate Research Assistant

Table 3.2.

SUMMARY OF TALKS[a] BASED, IN PART, ON OSR-GRANT

Date	Host Organization	Location	Topic(s)
12/4/85	Univ. Provence-Center for Dynamics/Thermodynamics of Fluids	Marseilles, France	2.1, 2.2
12/18/85	Univ. Bologna-Dept. ChE	Bologna, Italy	2.2
12/19/85	Tech. Univ. Milan-Dept. Aero.Engng	Milan, Italy	2.2
06/16/86	Stanford Univ./OSR Contractors Mtg	Palo Alto, CA	2.1, 2.2
06/19/86	Stanford Univ./OSR Contractors Mtg	Palo Alto, CA	2.3
07/10/86	American Scientist (Sigma Xi)	New Haven, CT	2.3
07/01--15/86	NATO Summer School on PCH: Interfacial Phenomena[c]	La Rabida (Huelva) Spain	2.1, 2.2
11/05/86	Amer. Inst. ChE.-National Mtg	Miami, FL	2.1
11/25/86	Princeton Univ. Depts. Aero/ME/ChE	Princeton, NJ	2.1, 2.2
12/16/86	Combustion Inst.-Eastern States Section[b]	San Juan, Puerto Rico	2.3

[a] Presented by D.E. Rosner (unless otherwise specified)
[b] Presented by Dr. A. Gomez (See Ref. 24)
[c] Presented by Dr. J. Castillo; See also Ref. (25).

4. CONCLUSIONS, FUTURE RESEARCH

In the OSR-sponsored research briefly described here we have shown that new laser-based experimental techniques for rapidly measuring vapor- and particle-mass transfer rates (1, 5, 14, 21), combined with recent advances in what might be called "thermophoretic boundary layer theories" (2, 3, 6-9, 19), are providing useful means to incorporate these phenomena in many propulsion engineering design/optimization calculations. In the future we hope to extend this work to include, among other things, the potentially important effects of turbulence in two-phase flows at high local particle mass loading (16), non-negligible particle inertia, and highly nonspherical particles (or molecules) (15). To shed light on boron particle ignition, "steady" combustion and extinction, our current research on the kinetics of boron gasification using MIPES is being extended to examine the $B_2O_3(g)/B(s)$ reaction probability and the response of such surfaces to imposed changes in temperature and reactant partial pressures.

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